IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

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| In re application of | : | |
| Eric Benazzi et al. | : | Group Art Unit: 1764 |
| Serial No. 10/018,526 | : | Examiner: GRIFFIN, WALTER DEAN |
| Filed: July 18, 2002 | : | |

For : FLEXIBLE METHOD FOR PRODUCTION OIL BASES WITH A ZSM-48 ZEOLITE

**DECLARATION UNDER 37 C.F.R. § 1.132**

**Honorable Commissioner**
**of Patent and Trademarks**
**Washington, D.C. 20231**

Sir :

I, Germain Martino, duly warned, declare and say as follows:

THAT, I am a French citizen; that I graduated from "Faculté des Sciences de l'Université de Strasbourg" (France) in 1961; that I obtained an Engineer Diploma from "Ecole Nationale Supérieure de Pétrole et des Moteurs" Rueil-Malmaison (France) in 1963; that I was received as a Doctor by "Université de Louvain" (Belgium) in 1965; and that I now reside in 78300 Poissy (France), 80 avenue Fernand-Lefebvre;

THAT, I was hired by "Institut Français du Pétrole" Rueil-Malmaison (France) in their Research Department to research on catalytic agents and catalytic reactions in May 1967; that, from January 1985 to September 1989, I was Manager of the Kinetics and Catalysis Research Division; that, from September 1989 to December 1997, I was Assistant Manager of the whole Refining and Petrochemical Technology Business Unit; and that since then I have been Manager of said Refining and Petrochemical Technology Business Unit.

I declare further:

THAT, I have supervised the following examples, and that these examples are correct.

**Example 1 (In accordance with the Invention)**

This example illustrates the advantages regarding the quality of the lubricating oil produced by the process of the invention, and regarding the improvements in respect to the yield, conversion and selectivity performed by said process.

The hydrocarbon feedstock used was a vacuum distillate " " Arabian Heavy (d15/4 =0.94, sulfur content of 3 % by weight, nitrogen content of 1200 ppm by weight) and have 97 % by weight of its components that boils above 370 °C. After dewaxing with a solvent MIBK (Methylisobutylcetone) at 20 °C, the viscosity index of this feedstock was equal to 50.

Firstly, the feedstock was sent to an hydrotreatment step achieved at a temperature of 380 °C, a pressure of 14 Mpa, a spatial velocity of 0.75 $h^{-1}$, in the presence of 1000 liters of hydrogen per liter of hydrocarbons, and in the presence of an amorphous catalyst comprising alumina, 3.6 % by weight of nickel (oxide), 17.2 % by weight of molybdenum (oxide) and 4 % by weight of phosphor (oxide) in order to hydrogen aromatics and nitrogen components, and in order to optimize the run of the hydrocracking catalyst downstream to the hydrotreatment catalyst. The 370°C$^+$ fraction conversion of the vacuum distillate produced from this hydrotreatment step was 30 % by weight.

The effluent of the hydrotreatment step was then sent to a hydrocracking step, without any intermediate separation step, the hydrocracking being achieved at a temperature of 370 °C, a pressure of 14 Mpa, a spatial velocity of 1.5 $h^{-1}$, in the presence of hydrogen, and in the presence of a catalyst comprising alumina, nickel, molybdenum, phosphor and a zeolite Y. The 370°C$^+$ fraction conversion of the vacuum distillate produced from this hydrotreatment step was 80 % by weight.

By atmospheric distillation of the effluent from the hydrocracking step in order to separate gas from liquid, a liquid fraction containing components having a boiling point above 370°C was obtained. This oil fraction presented a pour point of 39 °C and, after dewaxing with solvent MIBK at –20°C, a viscosity index of 130.

This liquid fraction was then directly treated by catalyst dewaxing at a temperature of 315 °C, a pressure of 14 MPa, a spatial velocity of 1 $h^{-1}$, in the presence of 1000 liters of hydrogen per liter of feedstock, and in the presence of a catalyst comprising a support based on 65 % by weight of a ZSM-48 zeolite (cf. paper on the magazine "zeolites", Vol. 5, 355 (185)) having a Si/Al ratio of 51 and a sodium content of 29 ppm, and on 35 % by weight of an alumina gel. The catalyst support was impregnated (by incipient impregnation) with an aqueous solution of $H_2PtCl_6$ and the platinum content of said support was 0.5 % by weight. The yield of dewaxed lubricant oil having components boiling above 370 °C was 82 % by

weight. This fraction presented a pour point of –21 °C and a viscosity index of 129. The yield of the " " middle distillate cut was 11% by weight.

All the effluents from the catalyst dewaxing step were then directly sent to a hydrofinishing step achieved at a temperature of 240 °C (difference between the dewaxing temperature and the hydrofinishing temperature equal to 75 °C), a pressure of 14 MPa, a spatial velocity of 0.5 $h^{-1}$, in the presence of 1000 liters of hydrogen per liter of feedstock, and in the presence of an amorphous aromatic hydrogenation catalyst having 0.6 % by weight of platinum deposited on an alumina matrix. During the hydrofinishing step, because of the low temperature, the cracking of the lubricant oil fraction was insignificant. The yield of the 370°C$^+$ fraction was above 99 % by weight. Besides, because of the low temperature of the hydrofinishing, most of the aromatics were hydrogenated which allows the production of medicinal type white lubricant oil.

The effluent from the hydrofinishing step was then sent to a distillation step comprising an atmospheric distillation in order to produce a lubricant oil fraction and a " " middle distillate cut having the properties presented in the table below :

| **Properties** | **Oil fraction 370°C$^+$** | **Middle distillate cut 150-370°C** |
|---|---|---|
| Yield to the HDF feed | 99 | - |
| D15/4 density | 0.845 | 0.827 |
| Sulfur content, ppm poids | <1 | <1 |
| Viscosity at 100°C | 5.9 | - |
| Point d'écoulement, °CPour Point | -21 | - |
| Indice de Viscosité Viscosity Index | 129 | - |
| Cétane ASTM D613 Cetane Number ASTM D613 | - | >60 |
| Aromatics by the Burdett method (UV), % poids | <0.1 | <0.1 |
| Aromatics by UV absorption ASTM D2008 | | |
| 260-280 nm | 0.0018 | - |
| 280-290 nm | 0.0016 | - |
| 290-299 nm | 0.0003 | - |
| 300-359 nm | 0.0002 | - |
| 360-400 nm | 0.0000 | - |
| 300-329 nm | 0.0002 | - |

| | | |
|---|---|---|
| 330-350 nm | 0.0001 | - |
| Aromatics by UV absorption on pure product (cell of 1 cm) 275 nm | 1.05 | - |
| Readily Carbonizable Substances | Acceptable | - |
| Saybolt color | +30 | - |

The steps of the process of the invention allow to considerably increase the viscosity index of the 370°C$^+$ lubricant oil fraction and to obtain a pour point in accordance to the current oil bases specifications. The temperature of hydrofinishing step of the process of the invention allows to produce a lubricant oil having a very low aromatic compounds content.

**Example 2 (comparative)**

The same feedstock as used in Example 1 was submitted to hydrotreatment, hydrocracking and atmospheric distillation steps, using the same operating conditions than in Example 1.

The 370°C$^+$ fraction obtained after distillation was then sent to a dewaxing step using the same catalyst and the same operating conditions than in Example 1.

The dewaxed effluent was then directly sent to a hydrofinishing step achieved at a temperature of 300 °C (difference between the dewaxing temperature and the hydrofinishing temperature being equal to 15 °C), a pressure of 14 MPa, a spatial velocity of 0.5 $h^{-1}$, in the presence of 1000 liters of hydrogen per liter of feedstock, and in the presence of the same hydrogenation catalyst as used in Example 1. During the hydrofinishing step, the 370°C$^+$ lubricant oil fraction yield was less by comparison to Example 1 (98 % by weight instead of 99 % by weight).

The effluent from the hydrofinishing step was then sent to a distillation step comprising an atmospheric distillation in order to produce a lubricant oil fraction and a middle distillate cut having the properties presented in the table below :

| Properties | Oil fraction 370°C+ | Middle distillate cut 150-370°C |
|---|---|---|
| Yield to the HDF feed | 98 | - |
| D15/4 density | 0.848 | 0.828 |
| Sulfur content, ppm poids | <1 | <1 |
| Viscosity at 100°C | 6.3 | - |
| Point d'écoulement, °CPour Point | -21 | - |
| Indice de Viscosité Viscosity Index | 126 | - |
| Cétane ASTM D613 Cetane Number ASTM D613 | | >60 |
| Aromatics by the Burdett method (UV), % poids | 1.5 | 1.0 |
| Aromatics by UV absorption ASTM D2008 | | |
| 260-280 nm | 0.0400 | - |
| 280-290 nm | 0.0343 | - |
| 290-299 nm | 0.0225 | - |
| 300-359 nm | 0.0177 | - |
| 360-400 nm | 0.0055 | - |
| 300-329 nm | 0.0177 | - |
| 330-350 nm | 0.0068 | - |
| Aromatics by UV absorption on pure product (cell of 1 cm) 275 nm | >10 | - |
| Readily Carbonizable Substances | Not acceptable | - |
| Saybolt color | +22 | - |

By comparison to Example 1, the higher temperature used at the hydrofinishing step in Example 2 (300 °C instead of 240 °C) led to the production of a lubricant oil containing more aromatic compounds with a lower yield.

The undersigned declares further that all statements are made herein of his own knowledge are true and that all statements made on information and belief are believed to be true ; and further that these statements are made with the knowledge that willful false statements and the like so made were punishable by fine or imprisonment, or both under Section 1001 Title 18 of United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Germain Martino

Rueil, October 13, 2004 Germain MARTINO